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Research on the purification effect of major pollutants in water by modular constructed wetlands with different filler combinations

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ABSTRACT

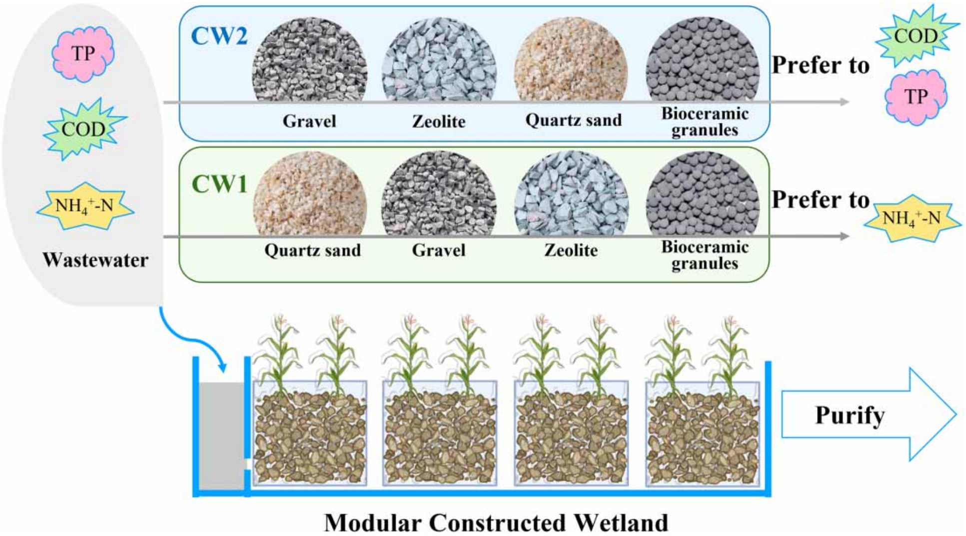
Constructed wetland systems have been widely used in China due to their advantages of good treatment effect, low cost and environmental friendliness. However, traditional constructed wetlands have challenges in application such as deactivation due to filler clogging, difficulty in filler replacement and low adaptability. To address the above problems, this research proposes a modular filler design constructed wetland based on the concept of assembly construction, which can quickly replace the clogged filler without destroying the overall structure of the wetland. Four commonly used fillers were selected and applied to the pilot system of the assembled constructed wetland in this study, in order to investigate the purification effect of the constructed wetland system with different filler module combinations (CW1, CW2, CW3) on the simulated wastewater. The results showed that the filler combination CW1 was the best for the removal of NH_4^+-N, and for TP and COD, CW2 has the best removal effect. Therefore, the assembled constructed wetland is adjustable and substantially reduces the maintenance cost, which provides technical guidance for its application in engineering.

Key words: blocking, filler combinations, modular constructed wetland, modularization, purification effect

HIGHLIGHTS

- Proposed a pilot system for the prefabricated constructed wetland.
- Explored the purification effects of different combinations of fillers.
- Analyzed concentration changes of pollutants along the route.
- Conducted cost analysis for replacing fill in two types of wetlands.

GRAPHICAL ABSTRACT



1. INTRODUCTION

With the continuous development of society and economy, the issue of water pollution and water safety has become an essential factor limiting global environmental safety and development. In recent years, various types of aquatic ecosystems have emerged as some of the most threatened ecosystems globally. Constructed wetland technology, which integrates wastewater treatment with ecological restoration, not only effectively addresses water pollution but also enhances environmental aesthetics, creating ecological landscapes that yield both environmental and economic benefits (Shukla *et al.* 2022). These systems, with their unique advantages, are gaining increasing attention and are being extensively applied in diverse sectors including urban wastewater treatment plant effluents, industrial wastewater, agricultural non-point source pollution (Duan & Feng 2022), rural decentralized domestic sewage, and mining and petroleum extraction wastewater treatment (Alalivi & Aslan 2022). In order to meet China's increasingly stringent requirements for surface water environments, the construction of constructed wetlands has played a crucial role in the field of wastewater treatment research in China.

According to the different ways of water intake, the constructed wetland is mainly divided into surface flow constructed wetland (Vymazal 2022), horizontal submerged flow constructed wetland and vertical submerged flow constructed wetland (Uthirakrishnan *et al.* 2022). In the horizontal submerged flow constructed wetland system, the direct contact between the effluent and the air is reduced due to the flow of effluent in the pores of the filler and the roots of the plants, and the hygienic conditions are better (Bydalek *et al.* 2023). In a study conducted by Zhou *et al.* (2021) in a pilot-scale surface flow constructed wetland (SFCW), the SFCW performed similarly or better in terms of removal efficiencies (REs) for salt, total nitrogen (TN) and total phosphorus (TP), even under high salinity conditions. Therefore, horizontal submerged constructed wetlands are widely used in engineering applications. Yet, constructed wetlands have certain limitations in practical application.

The long-term operation of constructed wetlands often leads to significant challenges such as clogging, reduced efficiency in nitrogen and phosphorus interception, and difficulties in regenerating fillers (Li *et al.* 2022). These issues have become a 'critical flaw' in the practical functioning of many constructed wetlands. Conventional remediation involves the extraction and replacement of clogged fillers, a process that can severely disrupt the overall structure of the wetland and result in extended system downtime. This highlights a crucial need for innovative solutions to enhance the sustainability and maintenance efficiency of constructed wetland systems (Keng *et al.* 2021). The concept of modular filler is to produce filler as an industrialized product in a large scale by means of a uniform structural form and then install it on site. The 'filler modular' constructed wetland can quickly replace the filler in the clogged area, thus effectively solving the clogging problem of the constructed wetland (Huang *et al.* 2022). In addition, assembled buildings have been used in various types of projects, such as residential and public facilities, due to their advantages of low construction cost, fast construction speed and low pollution, and have become the focus of China's construction industry. Applying the assembly concept to constructed wetlands can improve the construction efficiency, reduce the damage to the natural environment during the construction process, and increase the flexibility of constructed wetlands (Shi *et al.* 2023).

Fillers play an integral role in constructed wetlands, directly influencing the system's purification efficiency. Typically, fillers remove pollutants from raw water through a series of physicochemical processes, including physical interception, sedimentation, filtration, adsorption, and complexation (Xu *et al.* 2022a). Beyond acting as mediums for pollutant removal, fillers also support the growth of aquatic plants within the wetland, thereby enhancing ecological dynamics. Additionally, the surface of these fillers provides a habitat for microbial communities, facilitating biofilm formation (Dai *et al.* 2022). The diversity in the types of fillers, each with distinct physicochemical properties, results in varying degrees of purification effectiveness against different types of pollutants (Shukla *et al.* 2022). Consequently, selecting appropriate fillers based on the specific characteristics of the sewage is crucial to optimize purification outcomes and enhance the adaptability of constructed wetlands. Yang *et al.* (2022) has demonstrated that optimized combinations of fillers typically yield enhanced purification results, underscoring the importance of strategic filler selection in the design and operation of constructed wetlands. Hence, constructing several kinds of filler modules and matching and combining them can improve the flexibility and adaptability of constructed wetlands.

To address the problem that the conventional constructed wetland structure is very easy to be clogged, based on the design concept of an assembled building, this study innovatively designed a modular constructed wetland that is easy to replace the clogging filler. A pilot-scale modular constructed wetland model was then constructed. The purification effects of different filler modules on NH_4^+-N, TP and chemical oxygen demand (COD) in sewage treated by the fabricated constructed wetland system were investigated under two different flow conditions: intermittent flow and continuous flow. Additionally, the concentration variations of NH_4^+-N, TP and COD in the sewage were measured throughout the modular wetland system to analyze the underlying purification mechanisms. This approach represents a novel solution to traditional wetland system limitations, offering insights into the optimization of sewage treatment processes within a modular constructed wetland framework.

2. MATERIALS AND METHODS

2.1. Influent water quality

The advanced treatment of sewage plant effluents involves further removal of residual pollutants from primary and secondary treated water to meet the quality requirements for either reuse or discharge into receiving water bodies. Accordingly, in this study, the influent concentration of the main pollutants in the modular constructed wetland system was simulated based on the Level A Standards of the 'Pollutant Discharge Standard of Municipal Wastewater Treatment Plant' (GB 18918-2002). A large proportion of the organic matter in wastewater is microbially degradable, and glucose is a simple sugar that can be used as a representative of COD. Both NH_4Cl and KH_2PO_4 are commonly used in agricultural fertilisers, and their compositions are close to those of ammonia nitrogen and phosphate found in actual wastewater, and they are commonly used to simulate ammonia nitrogen and phosphorus in wastewater. The simulated wastewater used in this research was prepared by adding glucose, NH_4Cl and KH_2PO_4 to tap water, with the concentrations of the main pollutants as detailed in Table 1. This approach facilitates an understanding of the wetland system's efficacy in purifying wastewater to meet stringent environmental standards.

2.2. Design of filler modules

The filler modules in the prefabricated constructed wetland are composed of custom stainless steel filler cages, wetland fill materials and aquatic iris plants. The schematic representation of their assembly is illustrated in Figure 1.

Table 1 | Water quality parameters

Index	**COD (mg/L)**	**NH_4^+–N (mg/L)**	**TP (mg/L)**	**pH**
GB 18918-2002 (Level A Standard)	50.0	5.0 (8.0)	0.5	–
Concentrations	50.0	8.0	1.0	7.3 (±0.2)

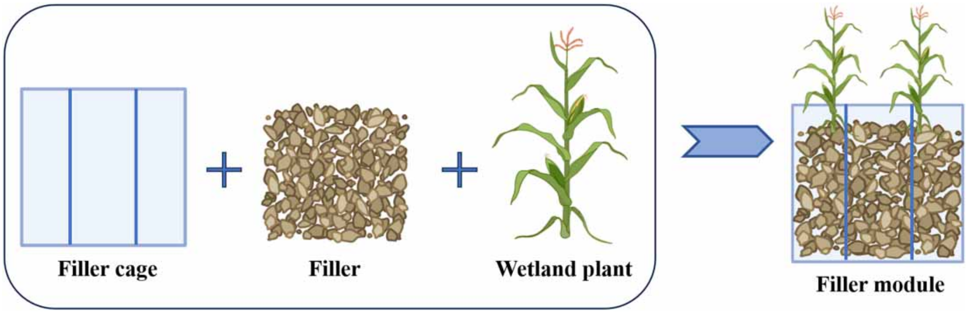


Figure 1 | Schematic diagram of the filler module composition.

2.2.1. Filler cages and wetland plants in the pilot system

The pilot system is equipped with four filler cages, each constructed from stainless steel wire mesh, with dimensions L × W × H = 0.3 m × 0.3 m × 0.3 m. Handles are provided on two sides of each cage for ease of module replacement and transportation. The mesh size on the sides of the cages is 3.0 mm × 3.0 mm, a design that prevents the escape of fill material while ensuring normal water flow. The sides of the cages are reinforced with steel bars to prevent deformation or breakage under the pressure of fill material and water flow.

Considering the influence of the climatic environment, wetland plants that are most suitable for constructed wetland projects in Central China primarily include reeds, iris, cattails, leeks and chive vegetables, water hyacinths and canna lilies. After careful selection and comparison, this study chose aquatic iris (*Iris pseudacorus*) as the wetland plant for the pilot model. The aquatic iris, sourced from a constructed wetland in Wuhan, Hubei Province, has an average height of 25 cm. Before planting, the roots were thoroughly washed to remove soil and then transferred to a nutrient solution for cultivation. The planting density in the wetland was 30 plants/m^2.

2.2.2. Fillers for constructed wetlands

Fillers that are commonly used in constructed wetlands can be categorized into three primary types based on their source: natural fillers, byproducts of industrial and agricultural processes, and artificially synthesized fillers. Natural fillers are materials that can be used in constructed wetlands with little or no processing, such as natural zeolite and gravel. Industrial and agricultural byproducts are residues that are generated during manufacturing and farming activities. One example of such a byproduct is quartz sand. Artificially synthesized fillers are those that have been industrially manufactured or processed to alter their inherent properties, creating novel materials such as bioceramic granules.

In this study, bioceramic granules, gravel, quartz sand and natural zeolite, which are commonly used in practical constructed wetland projects, were selected as the fillers for the different filler modules. Bioceramic granules feature a developed microporous structure with a large specific surface area, endowing them with excellent adsorption capabilities. Their microporous structure also facilitates microbial colonization and biofilm formation on their surface, which is beneficial for the biological degradation of pollutants (Goldberg *et al.* 2022). Gravel, known for its high hydraulic permeability, is widely available, non-toxic, harmless and cost-effective, making it frequently used as filler in constructed wetlands globally (Tanner & Sukias 1995). Quartz sand, which is characterized by its purity, resistance to compression and wear, high mechanical strength, stable chemical properties and strong interception capacity, has become the most extensively used and voluminous material in the water treatment industry (Wang *et al.* 2023b). Natural zeolite, with its unique structure, offers effective ion exchange capabilities and is commonly utilized in wastewater treatment (Ruiz-Ocampo *et al.* 2022). Bioceramic granules, gravel, quartz sand and natural zeolite were all procured from Gongyi HuiZhi Water Supply Materials Co., Ltd. (Gongyi City, China). The parameters for these four selected fillers are presented in Table 2.

2.3. Design and operation of the pilot constructed wetland system

2.3.1. Modular constructed wetland model

This study developed a horizontal subsurface flow constructed wetland model using custom-made organic glass. Based on preliminary experimental research, different filler modules were arranged in various sequences to investigate their

Table 2 | Parameters of the fillers

Fillers	Bioceramic granules	Gravel	Quartz sand	Zeolite
Size (mm)	7.00 (±1.00)	8.00 (±1.00)	7.00 (±1.00)	6.00 (±1.00)
Stacking density (g/cm^3)	1.13	1.61	1.75	0.76
Apparent density (g/cm^3)	1.94	2.62	2.66	1.68
Poriness (%)	41.35	39.00	43.00	52.00

purification effects on simulated wastewater from treatment plants. The location of the constructed wetland model is a water treatment laboratory in the School of Civil Engineering and Architecture, Wuhan University of Technology, Wuhan, China.

The schematic diagram of the pilot system is shown in Figure 2. The effective internal dimensions are 182 cm in length, 30.5 cm in width, and 30.5 cm in height, comprising an inflow area, four filler modules, inter-module sampling zones, and an outflow area. The inflow and outflow areas are each 15 cm long. Each filler module is 30.5 cm long with a filler height of 27 cm and an overlying water depth of 3 cm. The sampling zones between filler modules are 10 cm long. Baffles, 3 cm in height, are placed on either side of every sampling zone. These baffles can be adjusted in height using a fixed frame to alter the water flow between two filler modules. This design also facilitates water sampling after it has passed through each filler module, allowing for adjustments to the filler module combination for optimal effectiveness. Additionally, each sampling zone is equipped with an outlet at the bottom for water sampling and treated water recirculation.

2.3.2. Sequence of filler module combinations

In this study, several different combinations of filler modules were designed to analyze their effectiveness in nitrogen removal, phosphorus removal and COD reduction. Three distinct sequences of filler module combinations, namely CW1, CW2, and CW3, were set up for the experiment to compare the purification effects of different filler module sequences on the main pollutants in wastewater. The specific sequences of these combinations are presented in Table 3. This comparative approach aims to identify the most effective arrangement of modules for optimizing the treatment process in constructed wetland systems.

2.3.3. Operational modes of the model

The study investigated the purification effects of constructed wetlands on primary pollutants using two operational modes: intermittent and continuous. Following the guidelines of the 'Technical Specification for Wastewater Treatment Engineering in Constructed Wetlands' (HJ 2005-2010), during the intermittent operational phase, the prepared simulated wastewater was introduced into the pilot system. After achieving a hydraulic retention time of 2 days, samples were collected from each sampling port, and water quality indicators were measured. The system was then drained before commencing the next cycle, with a hydraulic loading rate of 0.15 m^3/(m^2·d). In the continuous operational phase, the hydraulic loading rate was

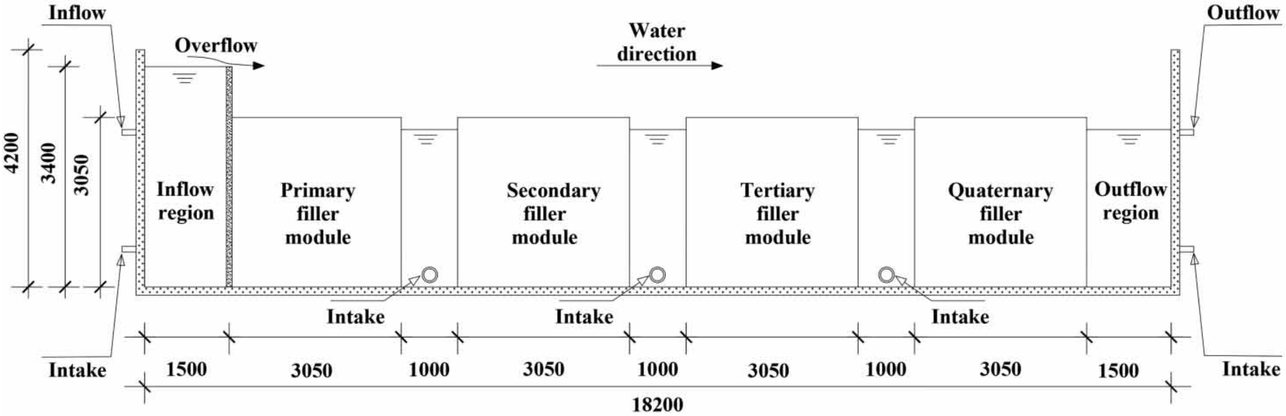


Figure 2 | Schematic diagram of the pilot system.

Table 3 | Different combinations of modular fillers

Number	Primary filler	Secondary filler	Tertiary filler	Quaternary filler
CW1	Quartz sand	Gravel	Zeolite	Bioceramic granules
CW2	Gravel	Zeolite	Quartz sand	Bioceramic granules
CW3	Zeolite	Gravel	Bioceramic granules	Quartz sand

maintained at 0.15 $m^3/(m^2{\cdot}d)$, which was identical to the intermittent flow. Water was drawn from the distribution tank using a peristaltic pump at a flow rate of 40 L/d to ensure continuous and uniform inflow. The raw water, after passing through the overflow port, sequentially entered different filler module groups before uniformly flowing out from the outflow area of the pilot system.

2.4. Chemicals and characterizations

For the preparation of simulated wastewater in this experiment, glucose, NH_4Cl and KH_2PO_4 were acquired from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China). Sodium hydroxide, potassium persulfate and potassium nitrate were sourced from Shanghai Yuan Ye Biological Technology Co., Ltd. (Shanghai, China). Permanganate, silver sulfate and mercuric sulfate were purchased from Fuchen (Tianjin) Chemical Reagents Co., Ltd. (Tianjin, China). Silver iodide, sulfanilic acid, potassium iodide and potassium sodium tartrate were obtained from Shanghai Macklin Biochemical Co., Ltd. (Shanghai, China). Molybdate, potassium antimonyl tartrate and ascorbic acid, all of analytical grade, were procured from Shanghai Jizhi Biochemical Technology Co., Ltd. (Shanghai, China). All solutions were prepared using deionized water. The ultraviolet spectrophotometer (UV-5100) used in this study was supplied by Shanghai Yuanxi Instrument Co., Ltd. (Shanghai, China). The COD-specific digestion instrument (H8049) was from Changsha Deko Instrument Equipment Co., Ltd. (Changsha, China). The peristaltic pump (Langer BT100-1F) for distribution was acquired from Jinan Ailabao Instrument Equipment Co., Ltd. (Jinan, China).

2.5. Detection methods

The measurement of $NH_4^+{-}N$ concentration was conducted using the Nessler's reagent spectrophotometric method. TP levels were determined through the ammonium molybdate spectrophotometric method. The COD was assessed using the potassium permanganate oxidation method (Rosario *et al.* 2023). pH levels were determined using a HACH DR2800 Multi-Parameter Water Quality Analyzer.

3. RESULTS AND DISCUSSION

3.1. Purification effect with different filler combinations of the constructed wetland

3.1.1. $NH_4^+{-}N$

This study explored the purification effect of CW1, CW2 and CW3 filler module combinations on $NH_4^+{-}N$ under intermittent flow and continuous flow operating conditions. In this case, the hydraulic retention time is 2 days, the hydraulic load is 0.15 $m^3/(m^2{\cdot}d)$ and the flow rate of the peristaltic pump is 40 L/d. The system is run eight times in a row, and the experimental data are recorded for each time. The results are shown in Figure 3.

Figure 3(a) illustrates that during the initial phase of intermittent flow operation, the purification effect of the three filler module combinations on $NH_4^+{-}N$ increases with the number of runs. Consequently, the removal rate of $NH_4^+{-}N$ increases from 77.5 to 92.5%. The results of the analysis indicate that the early stage's effective $NH_4^+{-}N$ removal is caused, in part, by the filler's adsorption of the molecule and, in part, by the microorganisms' rapid reproduction stage and the biological impacts' ongoing strengthening (Wang *et al.* 2023a). The purification effect has somewhat decreased and then stabilized as the number of runs continues to increase. This should be because, when operating in the intermittent mode, the system's filler matrix gradually reaches saturation in the $NH_4^+{-}N$ adsorption process, weakening the $NH_4^+{-}N$ removal impact (Zhao *et al.* 2022). Out of all of them, $NH_4^+{-}N$ is more purified by the CW1 filler sequence than by CW2 or CW3. This could be because different fillers have distinct physical and chemical characteristics, which means that different filler module combinations purify contaminants differently.

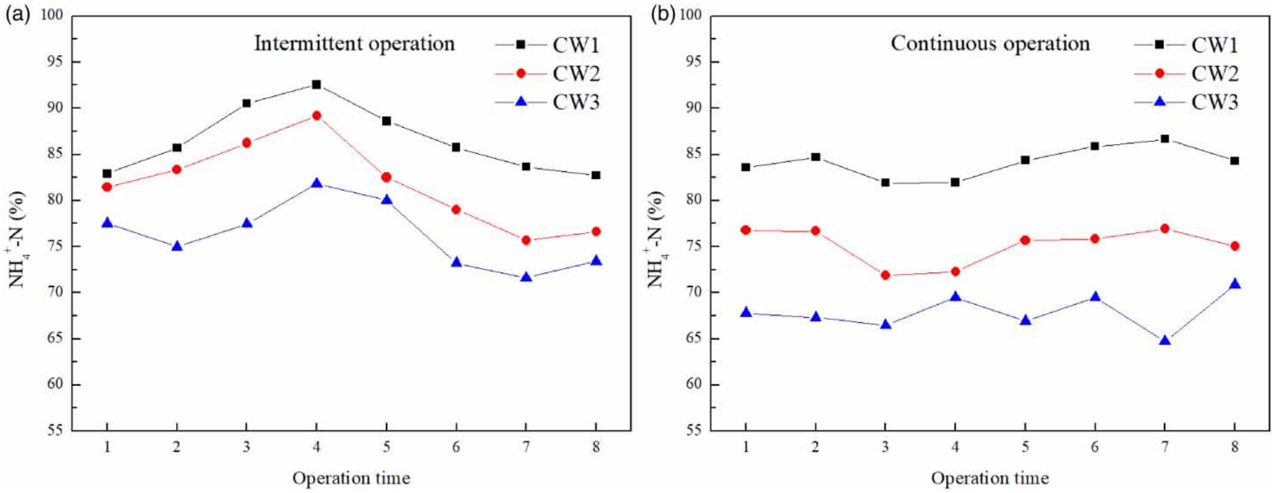


Figure 3 | Purification effect of modular constructed wetlands with different filler combinations on NH_4^+-N.

Figure 3(b) illustrates that the three combination sequences of CW1, CW2 and CW3 have a generally steady removal impact on NH_4^+-N under continuous flow operating conditions. Among them, CW1, CW2 and CW3 had average NH_4^+-N removal rates of 84.2, 75.1 and 65.9%, respectively, with notable variations. Better NH_4^+-N removal was also demonstrated by the CW1 filler sequence in continuous flow operation. To further investigate the causes, the NH_4^+-N concentration fluctuations in the wastewater along the modularly created wetland were next measured.

Analyzing Figure 3(a) and 3(b) together, it can be shown that the average removal rate of NH_4^+-N under the intermittent flow operating conditions of the three filler module combination sequences is better than it is under continuous flow operating settings. Among them, CW2 increased from 75.1 to 81.7%, CW3 from 65.9 to 76.3% and CW1 rose from 84.2 to 86.6%. The process by which nitrogen is removed from manmade wetlands involves the conversion of organic nitrogen by microorganisms into NH_4^+-N. This NH_4^+-N is subsequently oxidized to NO_3-N through a nitrification reaction, and finally, NO_3-N is reduced to N_2 or N_2O via a denitrification reaction and removed from the water. Among these, autotrophic aerobic microbes under aerobic conditions complete the nitrification reaction, while denitrifying bacteria under anaerobic conditions complete the denitrification reaction (Liu *et al.* 2023). The matrix filler is constantly submerged in water when water is flowing in continually. The wetland's limited capacity for natural reoxygenation means that the nitrifying bacteria in the system are unable to perform to the fullest extent of their abilities, which lowers the NH_4^+-N removal impact. Nevertheless, intermittent water inflow results in the wetland matrix being completely reoxygenated when it is not filled, which favorably enriches nitrifying bacteria. The ability of nitrifying and denitrifying bacteria to respond more effectively in anoxic and aerobic environments is advantageous for the removal of nitrogen. In addition, the operation mode of intermittent water intake provides construction conditions for the replacement of the filler module, so the intermittent water intake is more suitable for the wetland system.

3.1.2. Total phosphorus

This study explored the purification effect of CW1, CW2 and CW3 filler module combinations on TP under intermittent flow and continuous flow operating conditions. The results are shown in Figure 4.

Figure 4(a) illustrates that the CW2 filler sequence has the greatest removal effect on TP under intermittent flow operation conditions, with the highest removal rate reaching 86.7%. The next two had the highest removal rates, with CW1 and CW3 achieving 78.0 and 72.7%, respectively. Nevertheless, TP removal rates for CW1, CW2 and CW3 all exhibit a declining tendency as the number of runs rises. This is due to the fact that the primary methods of phosphorus removal in constructed wetlands are chemical precipitation and physical adsorption of fillers. On the one hand, while constructed wetlands continue to function, fillers' phosphorus absorption eventually reaches saturation (Li *et al.* 2023). Conversely, as the clogging process intensifies, the biofilm in the system steadily grows and the buildup of trapped substances in the spaces between the filler layers keeps growing, leading to a persistent decrease in the TP removal rate in the final phases. The TP removal rates of CW2, CW1 and CW3 decreased to 52.3, 32.5, and 37.6%, respectively, after the system was run eight times.

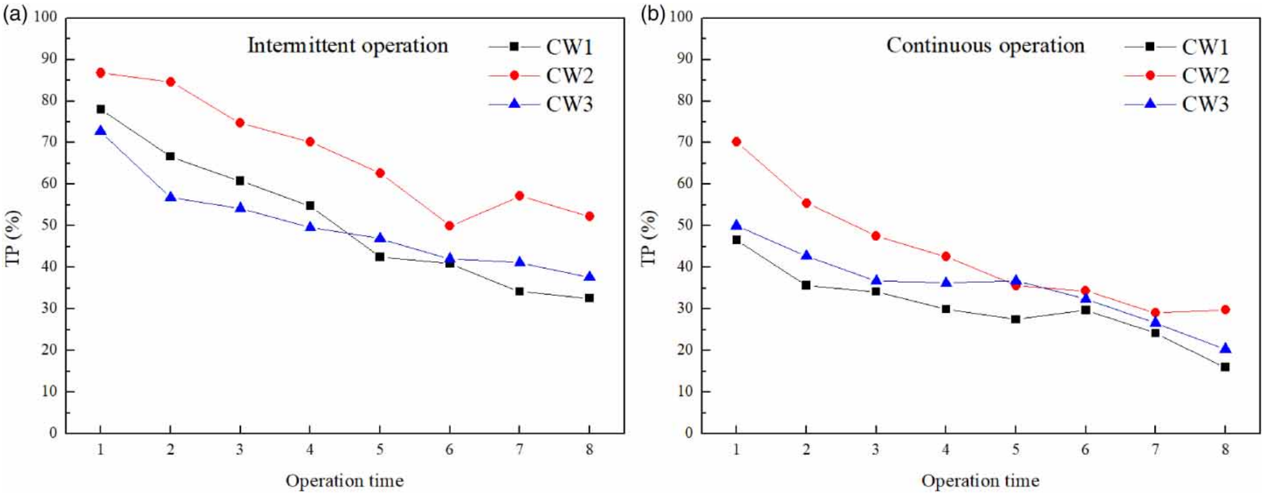


Figure 4 | Purification effect of modular constructed wetlands with different filler combinations on TP.

Figure 4(b) illustrates that the three filler module combination sequences have similar TP purification effects under continuous flow operation conditions, with CW2 sequence having the highest average removal rate of TP. It is noteworthy that the CW2 filler module combination sequence was demonstrated to be more effective in removing TP in both operating modes. In order to further examine the removal method, the concentration changes of TP following the wastewater's passage through several filler modules were measured.

3.1.3. Chemical oxygen demand

This study explored the purification effect of different combinations of CW1, CW2 and CW3 filler modules on COD under intermittent flow and continuous flow operating conditions. The results are shown in Figure 5.

As shown in Figure 5(a), under intermittent flow operating settings, the CW2 filler sequence has the best purifying impact on COD, reaching 89.8–94.9%, and the removal effect is reasonably constant. CW1 and CW3 have similar purifying effects on COD, with elimination rates reaching 66.6–85.0 and 62.4–85.7%, respectively. Figure 5(b) illustrates that, with a removal rate of 84.5–91.8%, CW2 continues to have the best COD removal performance under continuous flow operating conditions. The aforementioned experimental findings demonstrate that the two distinct constructed wetland operation modes have negligible effects on COD purification. This is so because the primary mechanism via which COD is removed is the

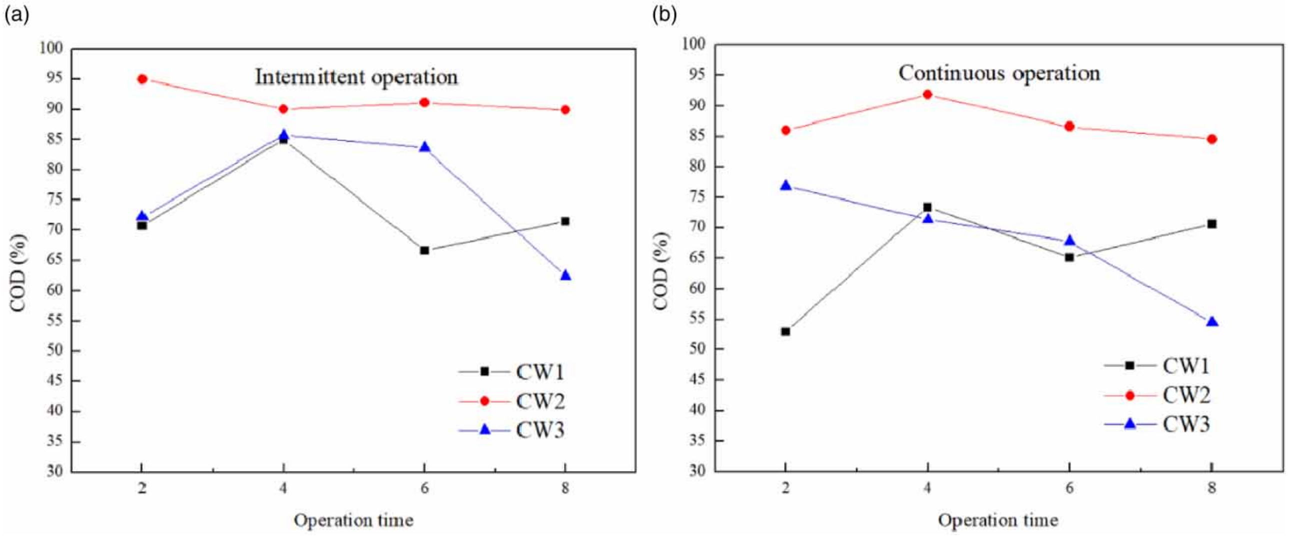


Figure 5 | Purification effect of modular constructed wetlands with different filler combinations on COD.

breakdown of bacteria in the biofilm that forms on the filler's surface. The organic content in the wastewater serves as nourishment for the microbes. Energy is created for the bacteria' own activities following degradation (Lu *et al.* 2021). Therefore, the water inlet method is not the primary factor influencing the COD purification effect.

Furthermore, the CW2 filler module combination sequence demonstrated superior COD elimination capabilities in both operating modes. The three filler modules' combination sequence affects COD purification in different ways. The reason for this could be that the system's many filler combination sequences enhance the variety of the microbiological life environment within the built-in wetland filler system. In order to do a thorough examination of its removal process, the concentration fluctuations of COD in wastewater along the modular created wetland were subsequently measured.

3.1.4. pH

From the purification mechanism of pollutants in constructed wetlands, it can be seen that pH will have a certain impact on the activity of microorganisms in wetlands and affect the adsorption of phosphorus by fillers, regardless of whether pH is too high, too low or greatly changed (Wang *et al.* 2022). Every sampling port's pH value was monitored constantly for the duration that the constructed wetland in this study was in operation. The test results are shown in Table 4. Studies conducted by Qian *et al.* (2019) demonstrate that the nitrification reaction necessitates the consumption of alkali throughout the denitrification phase of the created wetland system. When the nitrification reaction continues, the pH value will drastically decrease if the wastewater has insufficient alkalinity. However, nitrifying bacteria are highly sensitive to pH. When the pH is between 7.0 and 7.8, and between 7.7 and 8.1, respectively, nitrite and nitrifying bacteria are most active. Their activity rapidly decreases when the pH value rises above this range. It is worth noting that the pH values at the various sampling outlets in this study ranged from 7.03 to 8.09, indicating a more stable pH change in the constructed wetland. As a result, the pH level of the wastewater from the small test model of the constructed wetland used in this study has been kept within a somewhat ideal range, which promotes the system's effective operation.

3.2. Analysis of pollution removal along the route

In order to further explore its action principle, the changes in the concentrations of pollutants $NH_4^+{-}N$, TP and COD along the path of three filler module combinations (CW1, CW2 and CW3) were measured, and the average removal contribution rates of the corresponding filler modules were calculated, as shown in Table 5.

3.2.1. Concentration changes of $NH_4^+{-}N$ along the route and its analysis

During the operation of the pilot wetland system, samples were taken at the water inlet (0), between the filling modules (Samples 1, 2 and 3) and the water outlet (4), respectively, to explore the changes of $NH_4^+{-}N$ concentration in the system along the way, and the results are shown in Figure 6.

As shown in Figure 6(a), zeolite filler modules are placed far away from the water outlet for CW2 and CW3 under intermittent flow operation conditions. When the wastewater flows through the primary filler module, the concentration of $NH_4^+{-}N$ decreases rapidly. Lower concentration $NH_4^+{-}N$ wastewater then keeps passing through the secondary, tertiary and quaternary filler modules, and the rate of growth in $NH_4^+{-}N$ removal decreases. Of these, CW3 has the highest $NH_4^+{-}N$ removal rate (67.7%) of wastewater after it passes through the primary filler module (zeolite). This is because zeolite has exceptional ammonia nitrogen purification

Table 4 | Average pH of the water samples in the various intakes

CW1	**Quartz sand**	**Gravel**	**Zeolite**	**Bioceramic granules**
Intermittent operation	7.03	7.10	7.08	7.16
Continuous operation	7.51	7.36	7.39	7.28
CW2	**Gravel**	**Zeolite**	**Quartz sand**	**Bioceramic granules**
Intermittent operation	7.23	7.18	7.31	7.30
Continuous operation	7.64	7.82	7.89	7.71
CW3	**Zeolite**	**Gravel**	**Bioceramic granules**	**Quartz sand**
Intermittent operation	7.15	7.26	7.18	7.24
Continuous operation	7.86	7.96	8.09	7.93

Table 5 | Contribution rates of different filler module combinations to pollutant removal

NH_4^+-N removal contribution rate (%)	**CW1**	**Quartz sand**	**Gravel**	**Zeolite**	**Bioceramic granules**
	Intermittent operation	44.4	29.8	9.6	2.8
	Continuous operation	39.7	21.4	21.6	1.5
	CW2	**Gravel**	**Zeolite**	**Quartz sand**	**Bioceramic granules**
	Intermittent operation	63.8	7.1	2.9	7.8
	Continuous operation	21.2	31.2	18.7	4.0
	CW3	**Zeolite**	**Gravel**	**Bioceramic granules**	**Quartz sand**
	Intermittent operation	67.7	−0.9	5.0	4.4
	Continuous operation	48.7	9.0	9.0	−0.8
TP removal contribution rate (%)	**CW1**	**Quartz sand**	**Gravel**	**Zeolite**	**Bioceramic granules**
	Intermittent operation	19.9	1.9	13.2	16.2
	Continuous operation	5.7	10.7	4.9	9.1
	CW2	**Gravel**	**Zeolite**	**Quartz sand**	**Bioceramic granules**
	Intermittent operation	17.3	7.1	24.7	18.6
	Continuous operation	20.0	−2.7	10.0	14.8
	CW3	**Zeolite**	**Gravel**	**Bioceramic granules**	**Quartz sand**
	Intermittent operation	26.7	12.7	8.3	2.5
	Continuous operation	4.6	19.8	15.4	−3.7
COD removal contribution rate (%)	**CW1**	**Quartz sand**	**Gravel**	**Zeolite**	**Bioceramic granules**
	Intermittent operation	20.0	19.5	15.1	18.8
	Continuous operation	26.1	6.3	18.5	14.6
	CW2	**Gravel**	**Zeolite**	**Quartz sand**	**Bioceramic granules**
	Intermittent operation	57.1	17.4	9.7	7.2
	Continuous operation	39.3	23.8	17.6	6.5
	CW3	**Zeolite**	**Gravel**	**Bioceramic granules**	**Quartz sand**
	Intermittent operation	20.9	19.3	27.6	8.2
	Continuous operation	23.2	11.5	11.5	21.5

capabilities. However, lower concentration wastewater thereafter keeps passing through the gravel secondary filler, and an increase in the rate at which ammonia and nitrogen are removed from the water is observed to be negative. This may be due to the fact that, under the intermittent operation mode, gravel, which has a general ability to remove ammonia nitrogen, removes nitrogen from wastewater through the effects of adsorption and chemical complexation by the filler. When the gravel adsorption reaches saturation quickly, the adsorbed nitrogen is easy to be released again when the concentration of ammonia nitrogen in the influent water is low, resulting in an increase in the concentration of ammonia nitrogen. Ultimately, sampling port 4 of CW2 and CW3 had a removal rate of 76.2 and 81.7%, respectively. Compared to sampling port 1, the increase in removal rate is not significant. The 'weak' removal impact of the combined secondary, tertiary and quaternary fillers on wastewater with low NH_4^+-N content could be the cause.

The NH_4^+-N purifying effect for CW1 is superior to that of CW2 and CW3. When the initial wastewater with a high NH_4^+-N content flows through the primary filler module (quartz sand), 44.4% of NH_4^+-N is removed. After the wastewater continues to flow through the secondary filler module (gravel), the removal rate is also significantly increased (up to 74.2%), and the concentration of NH_4^+-N in the wastewater has been greatly reduced. Subsequently, after the wastewater with low ammonia nitrogen content flows through the tertiary filler module (zeolite), NH_4^+-N can still be largely removed, with the removal rate reaching 83.7% (9.2% higher than that of outlet 2). The excellent removal effect of zeolite on low concentration

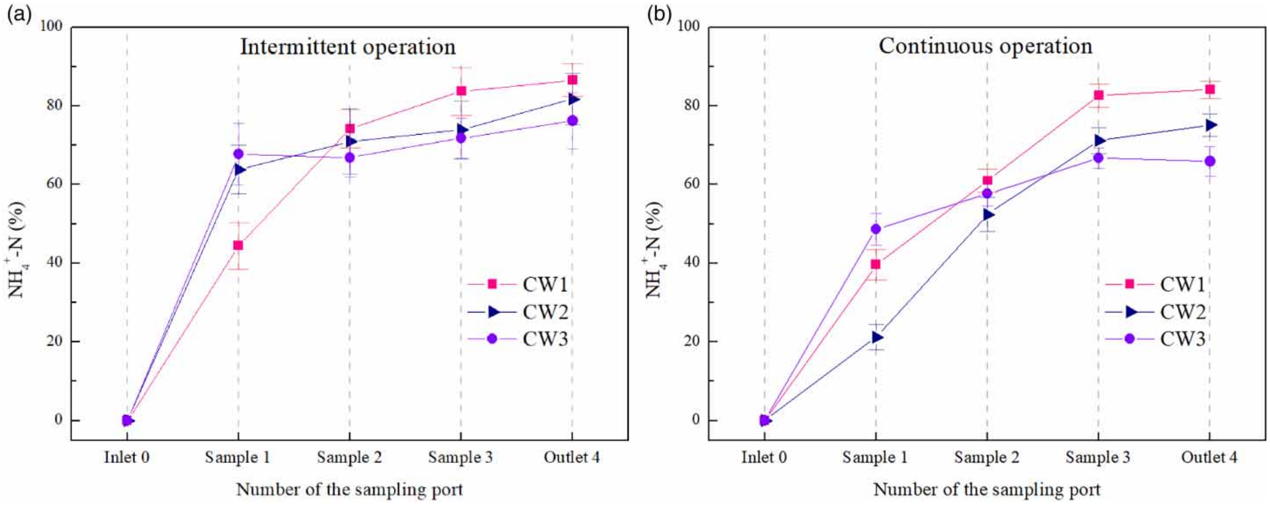


Figure 6 | The variations of NH_4^+-N removal along the process in intermittent (a) and continuous (b) processes.

ammonia nitrogen may be due to the characteristics of light weight, low resistance to water flow and not easy to clog. Moreover, the abundant pore structure on the surface of zeolite is easy to biofilm, so it is easier to form a living environment for nitrifying and denitrifying bacteria; therefore, the removal of ammonia nitrogen is particularly prominent, which is conducive to nitrogen removal (Cao *et al.* 2022). Finally, when the wastewater flows through the quaternary filler module (bioceramic granules), 2.8% of NH_4^+-N is removed, and the removal rate reaches 86.6%.

The comparatively constant influent flow rate and pollutant concentration in Figure 6(b) demonstrate how each stage filler has a specific removal impact on ammonia nitrogen under continuous flow operation conditions. CW1, CW2 and CW3 had average NH_4^+-N removal rates of 84.1, 75.1 and 65.9%, respectively, with notable variations. Furthermore, CW3 (zeolite) has the highest average removal rate of NH_4^+-N when wastewater flows through the primary filler, reaching 48.7%. CW1 (quartz sand) has the second-highest average removal rate, reaching 39.7%. On the other hand, CW2 (gravel, removal rate 21.2%) has the lowest average removal rate of NH_4^+-N. These findings are in line with Yang *et al.*'s (2022) research findings, which state that the removal effect of various fillers on NH_4^+-N is as follows: zeolite > quartz sand > gravel. It demonstrates once more how well zeolite removes nitrogen from ammonia. Therefore, zeolite filler with strong ammonia nitrogen removal capacity can be designed close to the water outflow to ensure that it also has a certain purification effect on wastewater with low ammonia nitrogen content, so ensuring the removal effect of the system on NH_4^+-N. In practical engineering applications, the removal effect of other pollutants should also be considered simultaneously.

3.2.2. Concentration changes of TP along the route and its analysis

The variations of TP removal in simulated wastewater by the combination of different filler modules (CW1, CW2 and CW3) are shown in Figure 7.

Figure 7(a) illustrates how different levels of TP removal occur when wastewater passes through different types of filler modules during intermittent flow operation. With an average removal rate of 67.7%, the CW2 filler module combination has the greatest TP removal rate among them. CW2 and CW3 had removal effects on TP that are equal – 51.3 and 50.1%, respectively. It is worth noting that under the filler combination sequence of CW1 and CW2, TP is significantly removed after wastewater flows through the quaternary filler module (bioceramic granules). During the experiment, it was found that a large amount of biological sediment was accumulated under the filler cage of bioceramic granules, which was mainly due to the developed microporous structure on the surface of biological ceramic, which was conducive to the membrane hanging and growth of microorganisms (Chen *et al.* 2019). Therefore, adsorption of sediment and filler is probably the primary form of phosphorus removal and the focus should be on cleaning the sediment underneath the bioceramic granules module when the filler module is replaced. In the actual engineering application, the bottom sediment at the bottom of the filler module can be removed periodically to maintain a good environment for phosphorus removal.

Figure 7(b) illustrates how, in comparison to intermittent operation, the TP removal impact is lessened during continuous flow operation, and at certain sampling ports, the TP concentration rises. For instance, the water's TP concentration rises as

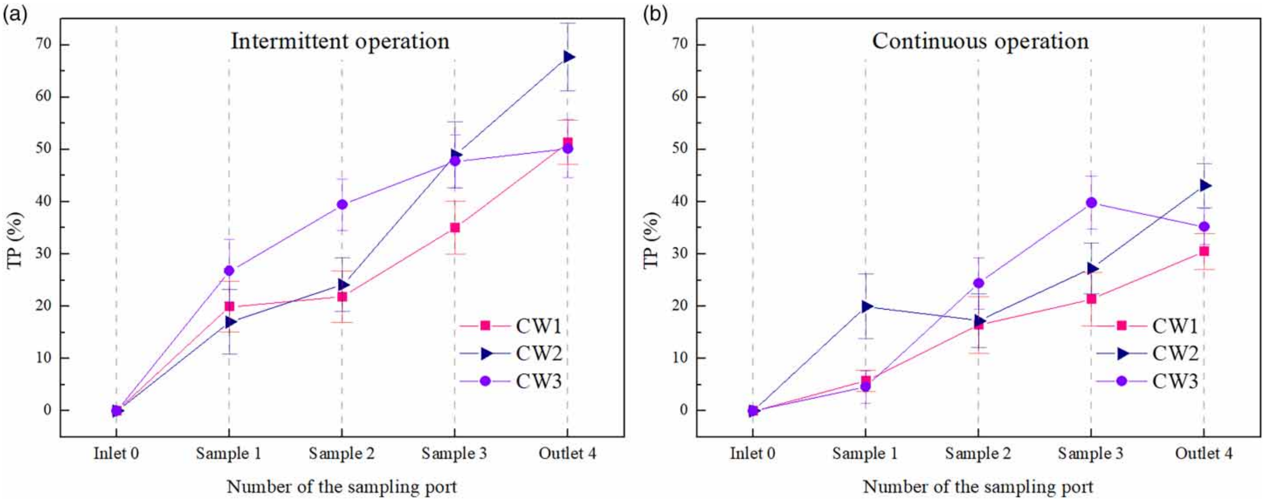


Figure 7 | The variations of TP removal along the process in intermittent (a) and continuous (b) processes.

wastewater passes through CW2's zeolite secondary filler. In CW3, the identical occurrence takes place. Wastewater passing through CW3's quartz sand quaternary filler reduces the TP removal rate from 39.8 to 35.2%. One possible explanation for this could be that the filler mostly removes phosphorous by physical adsorption, which eventually reaches saturation. Because microorganisms and plants do not remove as much TP, phosphorus release happens when the filler is saturated and adsorbed (Wang *et al.* 2012). It is worth noting that in the filler module combination sequence of CW1 and CW2, TP is greatly removed after wastewater flows through the quaternary filler module (bioceramic granules), which verifies that the bioceramic granules have a good purification effect on TP. Consequently, bioceramic granules can be added next to the water outlet when treating water bodies with high phosphorus content, which will help with TP removal and sediment collection.

3.2.3. Concentration changes of COD along the route and its analysis

The variations of COD removal in simulated wastewater by the combination of different filler modules (CW1, CW2 and CW3) are shown in Figure 8.

Figure 8(a) illustrates how, in the case of intermittent flow operation, different amounts of COD are eliminated when wastewater passes through each filler module. The COD removal rates of various fillers in the combination sequence of CW1 and

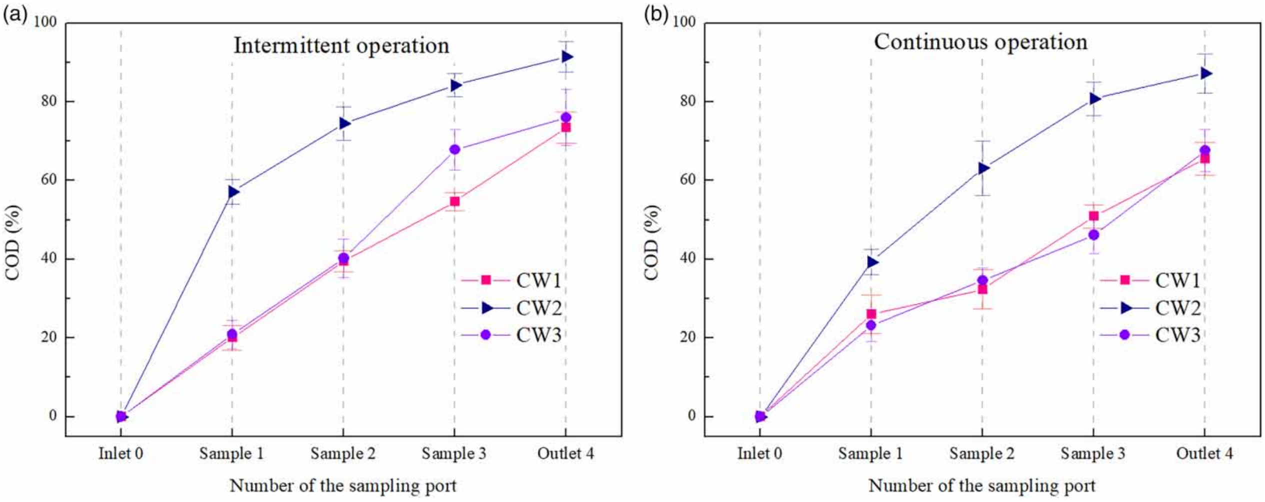


Figure 8 | The variations of COD removal along the process in intermittent (a) and continuous (b) operation.

CW3 are fairly balanced. Among them, the water effluent effects, which are 73.4 and 76.0%, respectively, are not significantly different. Compared to CW1 and CW3, CW2's sampling ports have greater removal rates overall, with an average removal rate of 91.5% for the final effluent. It is noteworthy that the purification impact of CW2 (gravel) is more pronounced and COD is eliminated at a rate of 57.1% when wastewater passes through the primary filler. This removal rate is comparable to that of CW1 (quartz sand) and CW3 (zeolite) 2.9 times. This demonstrates that gravel has a higher COD purifying impact. The main chemical composition of gravel is SiO_2 and Al_2O_3, which favors the rapid growth of aquatic plant roots due to its high hydraulic permeability coefficient. The well-developed root system provided a large number of attachment sites for microorganisms and COD was removed rapidly.

As can be seen from Figure 8(b), compared with intermittent flow, the COD removal effect is poor in continuous flow operation due to the poor reoxygenation capacity of the system. With an average removal rate of 87.2%, CW2 exhibits the best COD elimination effect. CW1 and CW3 had typical COD elimination rates of 65.5 and 67.6%, respectively. Similarly, CW2 (gravel, removal rate 39.3%) outperforms CW1 (quartz sand, removal rate 26.1%) and CW3 (zeolite, removal rate 23.2%) in terms of COD purification when wastewater passes through the primary filler module. This may be because, compared to zeolite and quartz sand, the pore structure of gravel filler helps to provide natural ventilation and oxygen transfer, easily creating a REDOX environment that encourages microbial REDOX reactions, and thus promotes the degradation of organic matter (Xu *et al.* 2022b). Furthermore, studies conducted by Cao *et al.* (2020) demonstrate that steady operation period of the created wetland system focuses on COD elimination in the area around the water inflow. Wastewater flow through the second part of the system is frequently constrained by factors like the quantity of dissolved oxygen and microorganisms present, and the RE of COD is low. Thus, in the constructed wetland system, adding gravel close to the wetland's water inlet can create a favorable redox environment and promote the development and reproduction of microorganisms within the wetland, maximizing their degradation effect and enhancing the wetland system's capacity to purify COD.

3.3. Filler replacement cost analysis

The fabricated constructed wetland in this study is a prefabricated and modular wetland treatment system that can provide an effective solution for replacing clogged fillers in engineering applications. Generally speaking, the fill replacement cycle of constructed wetlands is 5–10 years. This study takes a 100 m^3 constructed wetland as an example. After the wetland has been in operation for five years, the cost estimate of constructed wetland fill replacement is shown in Table 6.

As can be seen from Table 6, when replacing the filler in a traditional constructed wetland, the blocked filler in the wetland needs to be removed, and new filler must be purchased and filled. Therefore, the maintenance cost is relatively high, and the estimated cost is approximately 431,500 RMB. In addition, the constructed wetland needs to stop operation when replacing the filler, which has high environmental costs. However, the modular filler feature of prefabricated constructed wetlands makes the filler easy to install and replace. According to the condition of the filler and the operation of the wetland system, a filler replacement plan is formulated in advance, and the number, type and specification of the modules to be replaced, as well as the expected working time, can ensure the purification capacity and continuous operation of the wetland and reduce the cost of replacing the filler. Among them, the cost of replacing the filler of the prefabricated constructed wetland is about 255,000 RMB, which is about 40% lower than the cost of traditional constructed wetlands, and the economic

Table 6 | Cost estimation table for the filling replacement of 100 m^3 scale constructed wetland

Traditional constructed wetland				**Prefabricated constructed wetland**			
Cost type	**Quantity**	**Unit price (RMB)**	**Total price (RMB)**	**Cost type**	**Quantity**	**Unit price (RMB)**	**Total price (RMB)**
Wetland filler	77,000 kg	4.5/kg	346,500	No need for filler replacement			–
Labour cost	10 persons	3,000/person	30,000	Labour cost	10 persons	3,000/person	30,000
Machinery cost	1 time	50,000 /time	50,000	Machinery cost	2 times	50,000 /time	100,000
Transport cost	1 time	50,000 /time	5,000	Transport cost	5 times	50,000 /time	25,000
Filler washing	–	–	–	Filler washing	5 times	20,000 /time	100,000
Outage operation	7 days	High environmental costs exist		Outage operation	—	—	—
Total costs		431,500		Total costs		255,000	

benefits are significant. It is worth noting that the prefabricated constructed wetland does not require long-term water outage when replacing the filler and will not have a major impact on environmental safety and wetland operation.

4. CONCLUSIONS

In this study, the design of the constructed wetland with modular filler was proposed based on the concept of assembled construction, and four commonly used fillers, namely bioceramic granules, gravel, quartz sand and zeolite, were selected to fill different module groups, so as to construct an assembled constructed wetland pilot system that is easy to install and transport and can be replaced with clogged filler quickly. The purification effects of different filler module combinations (CW1, CW2 and CW3) on NH_4^+-N, TP and COD in simulated sewage were investigated under intermittent flow and continuous flow operation conditions, respectively. Among them, CW1 (quartz sand–gravel–zeolite–bioceramic granules) was the most effective for the removal of NH_4^+-N in the effluent, and CW2 (gravel–zeolite–quartz sand–bioceramic granules) was the most effective for the removal of TP and COD in the effluent. When replacing the clogged filler, the cost of replacing the filler in the assembled constructed wetland was reduced by about 40% compared to the traditional constructed wetland. This study provides a new strategy for the constructed wetland filler clogging problem, and provides data support for the adjustability of the assembled constructed wetland itself, which provides technical guidance in its engineering application.

AUTHOR CONTRIBUTIONS

Xiaoting Liu contributed to conceptualization, validation, and editing. Xuhao Li was involved in conceptualization, writing the original draft, validation, and formal analysis. Xiangling Zhang was responsible for methodology, supervision, and reviewing and editing the writing. Shilong Cao contributed to the development of software. Rang Liu was involved in validating the work. Hui Zhao conducted the investigation. Hao Zhu provided resources. Chen Wang was responsible for data curation. Xinlu Xiao was responsible for visualization.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICTS OF INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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